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A model to predict the effective permittivity of heterogeneous multiphase structures with new bounds

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ABSTRACT

A great number of numerical and theoretical models have been proposed, by several authors to estimate the effective permittivity of heterogeneous multiphase structures. The physical effective properties of both composite and nanocomposite materials are known for its depending on the composition and the arrangement of its components. Until now the existing literature is not able to take into account the effect of the geometry except the effect introduced via the depolarization factor presented by some models, which depends on the inclusion geometry and which is known only for a limited number of inclusions, with classical shapes like the disc or some regular equilateral polygons. This issue presents a limitation to study many systems with non-regular geometries. In this paper, a new model, based on the association of capacitors is presented, gives us two new bounds of the effective permittivity. The new results are compared with the existing models and the results obtained by applying the finite element method (FEM). The results of the present model are shown in good agreement with the existing data. Moreover, the new model improves considerably the calculation time compared with the finite element method.

1. Introduction

Owing to their wide ranging applications, considerable research efforts have been devoted to the study of effective permittivity, for different heterogeneous multiphase shapes and internal topology of the component's structure [1,2]. Undoubtedly, the dielectric properties are depending mainly on the dielectric shape characteristics, the arrangement of components and their volume fraction [3].

During the last few decades an intensive literature has been proposed to predict the effective permittivity for a mixture of heterogeneous materials. The improvement and implementation of computer modeling methods have been proposed to predict the physical properties of hetero-structures from their chemical structure [4] and the volume fraction of components.

These properties play a central role in the development of materials science. Recently, A. Prasad and K. Prasad studied the effective permittivity of random composite media [5] they compared Cuming [6], Maxwell–Wagner [7], Webmann [8], and Skipetrov [9] equations yielded equivalent results and consequently they have been combined together and calculated as a single equation named MWWS. This study revealed that the Cuming equation has the highest degree of acceptability in all the cases. The mixture was a two-component mixture with a homogeneous background in which circles of other material were embedded in random positions, and the circles were allowed to touch and overlap, which means that the mixture also contained more complex cluster geometries than the 2-D spheres. Not so far, Serdyuk et al. [10] studied the numerical simulations of dielectric properties of composite material with periodic structure they found that the distributions of the micro-scale electric field and dielectric losses in the volume of the composite material are strongly dependent on the frequency, the same effect is also analyzed using classical models for composite dielectric structures. Myroshnychenko and Brosseau [11] studied the effective permittivity in a percolation composite, for this they used finite element calculations and they found a quantitative test of McLachlan (TEPPE) equation, by comparing its prediction of the effective permittivity to the simulation results obtained on systems with overlapping disks.

Nonetheless, these theories estimate the effective permittivity of dielectric mixture for specific cases of shape, arrangement and the nature of inclusion and host matrix. The simplest and probably the most common law to predict the effective permittivity is the theory of Maxwell Garnett [12], which is the most used model to compare the numerical simulations and experimental data. Those comparisons told us that the MG theory is uncertain for a range of volume fraction for some values of contrast and the inclusion

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type. Some other models exist too, and can be used to predict the effective permittivity where the conditions of applying the model were satisfied.

The main purpose of this paper is to expose a new model without using the depolarization factor; this model gives us two new bounds to determine the effective permittivity for periodic or random multiphase heterogeneous structures. To validate the new results, many applications are studied using different geometries of inclusion and the contrast ($k = \varepsilon_{inc}/\varepsilon_{mat}$) between the components, then, the results are compared to the well-known Maxwell Garnett theory and finite element method [13]. Moreover, the results of the present model are shown in good agreement with the existing data.

2. The mixing rules and effective approach models

It is more evident to think about effective medium approach MG theory and similar models [5]. Thus, a presentation of a composite material illustrates the homogenization of a multiphase mixture of different inclusions characterized by theirs relative permittivity $\varepsilon_{inc(i)}$ and shapes immersed in a host matrix with relative permittivity ε_{mat} (see Fig. 1).

The effective permittivity (ε_{eff}) is a complex function which depends highly on the inclusions arrangement, theirs shapes, theirs relative permittivity (then the contrast k), the volume fraction of each different inclusion and the interactions between them which occurs in high concentrations. Some of the existing models do not take into account one or more of those dependencies which explain the divergence observed between the existing models and the experimental data or the simulation results. So, the scientists often setup some parameters of a model by fitting their data or sometimes they modify the composite experimentally as done at [14] where a better correlation between experimental and analytical data can be observed when the epoxy matrix was modified with lower volume fraction of hollow ceramic spheres (20%) and finally, the epoxy– E-glass composite system can be used to obtain material with the exact values of dielectric constant and loss tangent [15].

The appliance of the effective medium approach is only possible when the quasi-static limit is verified otherwise, when the typical size of inclusions is small compared to the wavelength of the electromagnetic wave probing the heterogeneous structure.

The Maxwell Garnett equation was based on the induced polarization by a uniform external electric field applied to the spherical inclusions placed in the host matrix. For the 2D systems, the effective permittivity ε_{MG} for a biphasic heterogeneous structure with all inclusions have the same value of permittivity ε_{inc} is given by the following expression.

$$\varepsilon_{MG} = \varepsilon_{mat} + 2\phi_{inc}\varepsilon_{mat}\frac{(\varepsilon_{inc} - \varepsilon_{mat})}{\varepsilon_{inc} + \varepsilon_{mat} - \phi_{inc}(\varepsilon_{inc} - \varepsilon_{mat})} \tag{1}$$

where Φ_{inc}, ε_{inc} and ε_{mat} denotes the surface fraction of the inclusion, the relative permittivity of the inclusion and the relative permittivity of the host matrix, respectively.

A second other model was proposed by Looyenga [16] where the effective permittivity of the mixture is given by the following expression:

$$\varepsilon_{Lyng}^{1/3} = (1 - \phi_{inc}) \cdot \varepsilon_{mat}^{1/3} + \phi_{inc} \cdot \varepsilon_{inc}^{1/3} \tag{2}$$

Another interesting model was proposed by Wiener (1912)[17] and yet is used in many papers until writing those lines [18] and which is based on the association of two capacitors. Thus, two formulas were used, one for the parallel case given by the Eq. (3) and present the exact effective permittivity of the heterogeneous structures shown in the Fig. 2a, the second formula is used to find the effective permittivity of two capacitors mounted in series given by the Eq. (4) and presents the exact effective permittivity of the heterogeneous structure shown at the Fig. 2b.

$$\varepsilon_{W-par} = (1 - \phi_{inc}) \cdot \varepsilon_{mat} + \phi_{inc} \cdot \varepsilon_{inc} \tag{3}$$

$$\varepsilon_{W-ort} = \left(\frac{1 - \phi_{inc}}{\varepsilon_{mat}} + \frac{\phi_{inc}}{\varepsilon_{inc}}\right)^{-1} \tag{4}$$

Furthermore, another interesting model can be found in the literature, in this model two bounds were established known as Hashin-Shtrikman bounds [19]. For the 2D systems, the bounds are given by the following expression:

$$\varepsilon_{HS,1} = \varepsilon_{mat} + \frac{\phi_{inc}}{\left(\frac{1}{\varepsilon_{inc} - \varepsilon_{mat}}\right)\left(\frac{1 - \phi_{inc}}{2 \cdot \varepsilon_{mat}}\right)} \tag{5}$$

And

$$\varepsilon_{HS,2} = \varepsilon_{inc} + \frac{1 - \phi_{inc}}{\left(\frac{1}{\varepsilon_{mat} - \varepsilon_{inc}}\right)\left(\frac{\phi_{inc}}{2 \cdot \varepsilon_{inc}}\right)} \tag{6}$$

Also, other models are based on association of capacitors exist too, not so far, Patil et al. [20] have calculated the effective permittivity basing on the capacitance of an appropriate equivalent circuit. The main task was to find the equivalent capacitance of the capacitor containing the inclusion. The model was applied to a spherical inclusion and can be extended to any other inclusion type, but the problem appears in the case of non regular geometries, which is too hard or impossible to find the equivalent capacitance by a mathematical analysis.

Over all those ideas, the Eqs. (3) and (4) attracted less attention, however, could be used to calculate the effective permittivity for a random inclusion domain for a given volume fraction. By the way, to evaluate the effective permittivity many numerical methods were used previously such as the finite elements method [21,22] and finite difference and finite difference time domain methods [23–26] in this paper only the FEM is used to validate the present model. This method is used in the present, due to its performances moreover, actually is the most used method to estimate the effective permittivity for the heterogeneous structures. However, this technique is very expensive in term of computation and time cost. Thus, in the present, all the structures are exposed to an identical difference of potential (V_1–V_2) imposed to the y-axis direction, with the same square edge "L".

Solving the problem analytically, means find the solution of the Laplace's equation

$$\nabla(\varepsilon_0\varepsilon_r)\nabla V = 0 \tag{7}$$

where ε_r, V and ε_0 are the local relative permittivity, the local potential and $\varepsilon_0 = 8.854\,187\,817 \cdot 10^{-12}$ F/m the vacuum permittivity constant respectively.

The electrostatic energy W given by the following expression:

$$W = \frac{1}{2}\varepsilon_0 \iiint \varepsilon(x,y,z)\left[\left(\frac{\partial V}{\partial x}\right)^2 + \left(\frac{\partial V}{\partial y}\right)^2 + \left(\frac{\partial V}{\partial z}\right)^2\right]dxdydz \tag{8}$$

For a cubic capacitor where all edges are equal to L, and when the potential difference (V_1–V_2) is applied in the y-axis direction, the previous expression of the electrostatic energy W, could be written as follows:

$$W = \frac{1}{2}\varepsilon_0\varepsilon_{eff}L(V_1 - V_2)^2 \tag{9}$$

The effective permittivity is evaluated by substituting the first expression of the electrostatic energy Eq. (8), into the second Eq. (9).